

CLAIMS

1. A method for preparing active or selective solid catalysts of inorganic or organometallic materials or mixtures thereof by selecting a certain number of solid catalysts of different chemical composition or different weight composition or different chemical and different weight composition and determination of essential catalyst properties, comprising arbitrarily or randomly newly structuring by means of stochastic methods the individual catalyst components or amounts of mass of the catalyst components or the catalyst components and amounts of mass in the best catalysts of the first generation with respect to activity or selectivity or activity and selectivity for a certain catalytic reaction, determining the activity or selectivity or activity and selectivity of the obtained catalysts of the second generation, again arbitrarily or randomly newly structuring by means of stochastic methods the individual catalyst components or amounts of mass of the catalyst components or the catalyst components and amounts of mass of a portion of the best catalysts of the second generation, determining the activity or selectivity or activity and selectivity of the obtained catalysts of the third generation, and continuing these steps of new structuring of the best catalysts of all generations and the property determination up to obtaining one or more catalysts with the desired properties for the specific catalytic reaction.

2. The method for preparing active or selective solid catalysts according to claim 1, wherein

(a) for a catalytic reaction a number n_1 of solid catalysts of the elements of the periodic table of

the elements (PTE) in the form of compounds of the formula (I)

$$(A_{a_1}^1 \dots A_{a_i}^i) - (B_{b_1}^1 \dots B_{b_j}^j) - (D_{d_1}^1 \dots D_{d_k}^k) - (T_{t_1}^1 \dots T_{t_l}^l) - O_p \quad (I)$$

are prepared, wherein $A^1 \dots A^i$ is a quantity i of different main components which are selected from the elements of the PTE, except trans uranium and noble gas elements, and the number i is between 1 and 10,

$B^1 \dots B^j$ is a quantity j of different minor components selected from the group of the elements

Li, Na, Ka, Mg, Ca, Sr, Ba, Y, La, Ti, Zr, V, Nb, Cr, Mn, Tc, Re, Fe, Ru, Os, Co, Rh, Ir, Ni, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, Al, Ga, In, C, Si, Sn, Pb, N, P, As, Sb, Bi, S, Se, Te, F, Cl, Ce and Nd, and the number j is between 1 and 10,

$D^1 \dots D^k$ is a quantity k of different doping elements which are selected from the group of the elements

Li, Na, Ka, Mg, Ca, Sr, Ba, Y, La, Ti, Zr, V, Nb, Cr, Mn, Tc, Re, Fe, Ru, Os, Co, Rh, Ir, Ni, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, Al, Ga, In, Si, Sn, Pb, N, P, As, Sb, Bi, S, Se, Te, F, Cl, Ce and Nd and the number k is between 1 and 10,

$T^1 \dots T^l$ is a quantity l of different support components which are comprised of oxides, carbonates, carbides, nitrides, borides of the elements Mg, Ca, Sr, Ba, La, Zr, Ce, Al, Si or a mixed phase of two or more thereof, and the number l is between 1 and 10, and O is oxygen,

$a_1 \dots a_i$ are identical or different mole fractions of 0 to 100 mole-% with the provision that the mole fractions $a_1 \dots a_i$ cannot all at the same time be 0,

$b_1 \dots b_j$ are mole fractions of 0 to 90 mole-%,

$d_1 \dots d_k$ are mole fractions of 0 to 10 mole-%,

$t_1 \dots t_l$ are mole fractions of 0 bis 99.99 mole-%,

p is a mole fraction of 0 to 75 mole-%, wherein the sum of all mole fractions $a_i + b_j + d_k + t_l$ may be not greater than 100 %, and

the number n_1 of catalysts with different quantitate composition or different chemical composition or

different weight and chemical compositions is in the range of 5 to 100,000;

(b) the activity or selectivity or activity and selectivity of the n_1 solid catalysts prepared according to (a) of the first generation is determined experimentally for a catalytic reaction in a reactor or in several reactors switched parallel;

(c) a number of 1 - 50 % is selected as number n_2 from the number n_1 of the catalysts of the first generation having the highest activities for a specific reaction or highest selectivities for the desired product or product mixture of the catalytic reaction or activity and selectivity;

(d) the catalyst components contained in the number n_2 of the catalysts with a pre-set probability W , which results for each of the components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ from the corresponding equations

$$W_A = \frac{1}{i \cdot n_2} \cdot 100\%, W_B = \frac{1}{j \cdot n_2} \cdot 100\%, W_D = \frac{1}{k \cdot n_2} \cdot 100\%, W_T = \frac{1}{l \cdot n_2} \cdot 100\%$$

are exchanged between two catalysts selected from the number n_2 with a probability of $W_{cat} = \frac{1}{n_2} \cdot 100\%$ or

that the amount of mass $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ of the catalyst components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ for some of the catalysts selected with a probability of $W_{cat} = \frac{1}{n_2} \cdot 100\%$

are varied in that new values for the mole fractions $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ are determined within the limits defined under (a); or that exchange and variation are performed;

in this way new catalysts of the general formula (I) with the meaning of A, B, D, T, a, b, d, and t and p

as defined under (a) are produced in a number y_2 which form the catalysts of the second generation;

(e) the activities or selectivities or activities and selectivities of the y_2 solid catalysts of the second generation are determined experimentally for the same specific reaction as in (b) in one or more reactors;

(f) a number of the n_3 catalysts of the second generation, having the highest activities for a specific reaction or highest selectivities for the desired product and product mixture or the activities and selectivities of all solid catalysts of the first and second generation, is selected, wherein the number n_3 is 1 to 50 % of the number n_1 ;

(g) the catalyst components contained in the number n_3 of the catalysts with a pre-set probability W , which results for each of the components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ from the corresponding equations

$$W_A = \frac{1}{i \cdot n_3} \cdot 100\%, W_B = \frac{1}{j \cdot n_3} \cdot 100\%, W_D = \frac{1}{k \cdot n_3} \cdot 100\%, W_T = \frac{1}{l \cdot n_3} \cdot 100\%$$

are exchanged between two catalysts selected from the number n_3 with a probability of $W_{cat} = \frac{1}{n_3} \cdot 100\%$ or

that the amount of mass $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ of the catalyst components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ for some of the catalysts selected with a probability of $W_{cat} = \frac{1}{n_3} \cdot 100\%$

are varied in that new values for the mole fractions $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ are determined within the limits defined under (a) or exchange and variation are performed;

in this way new catalysts of the general formula (I) with the meaning of A, B, D, T, a, b, d and t and p

as defined under (a) are produced in a number y_3 which form the catalysts of the third generation;

(h) the activity or selectivity or activity and selectivity is of the y_3 new solid catalysts of the third generation prepared according to (g) is determined experimentally for the same specific reaction as in (b) in one or more reactors;

(i) a number of n_{n+1} solid catalysts of the ^{first to} n -th generation, having the highest activities for a catalytic conversion or the highest selectivities for the desired product and product mixture or the highest activity and selectivity of all solid catalysts of the first to n -th generation, is selected, wherein the number n_{n+1} is 1 to 50 % of the number n_1 ;

(j) the catalyst components contained in the number n_{n+1} of the catalysts with a pre-set probability W , which results for each of the components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ from the corresponding equations

$$W_A = \frac{1}{i \cdot n_{n+1}} \cdot 100\%, W_B = \frac{1}{j \cdot n_{n+1}} \cdot 100\%, W_D = \frac{1}{k \cdot n_{n+1}} \cdot 100\%, W_T = \frac{1}{l \cdot n_{n+1}} \cdot 100\%$$

are exchanged between two catalysts selected from the number n_{n+1} with a probability of $W_{cat} = \frac{1}{n_{n+1}} \cdot 100\%$

or that the amount of mass $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ of the catalyst components $A^1 \dots A^i, B^1 \dots B^j, D^1 \dots D^k$ and $T^1 \dots T^l$ for some of the catalysts selected with a probability of $W_{cat} = \frac{1}{n_{n+1}} \cdot 100\%$

are varied in that new values for the mole fractions $a_1 \dots a_i, b_1 \dots b_j, d_1 \dots d_k$ and $t_1 \dots t_l$ are determined within the limits defined under (a), or that exchange and variation are performed;

in this way new catalysts of the general formula (I) with the meaning of A, B, D, T, a, b, d and t and p as defined under (a) are produced in a number y_{n+1} which form the catalysts of the (n+1)-th generation;

(k) the activity or selectivity or activity and selectivity of the y_{n+1} solid catalysts of the (n+1)-th generation prepared according to (g) is determined experimentally for the same specific reaction as in (b) in one or more reactors;

(l) the selection according to the steps (c) + (f) + (i), the preparation of a new catalyst generation according to the steps (d), (g), (j), and the activity/selectivity determination according to the steps (e) + (h) + (k) is continued up to obtaining a catalyst generation in which the activity or selectivity or activity and selectivity relative to the previous generations as an arithmetic mean is not increased or no longer significantly increased.

3. The method according to claim 2, wherein the exchange of the catalysts or the variation of the amount of mass or exchange and variation in the portions (d), (g), and (j) are carried out by means of a numerical random-check generator.

4. The method according to claim 3, wherein the program codes G05CAF, G05DYF, G05DZF and G05CCF of the NAG Library (NAG FORTRAN Workstation Library, NAG Group Ltd., 1986) of a numerical random-check generator are used.

A 5. The method according to claim 2(~~a~~), wherein the quantity n_1 is in the range of 5 to 100 for catalysts which are different with regard to their weight composition or chemical composition or weight and chemical composition.

6. The method according to claim 2, wherein the selection number n_2 , n_3 , or n_{n+1} is 5 to 30 % of the quantity n_1 .

7. The method according to claim 2, wherein the main components are selected from the group comprised of Mg, Ca, Sr, Ba, Y, La, Ti, Zr, V, Nb, Cr, Mn, Tc, Re, Fe, Ru, Os, Co, Rh, Ir, Ni, Pd, Pt, Cu, Ag, Au, Zn, Cd, Hg, B, Al, Ga, In, C, Si, Sn, Pb, N, P, As, Sb, Bi, S, Se, Te, F, Cl, Ce und Nd.
8. The method according to claim 2, wherein the mole fractions $b_1 \dots b_j$ are from 0 to 50 mole-%.
9. The method according to claim 2, wherein the preparation of the catalyst mixtures is carried out by mixing salt solutions of the elements of the components $A^1 \dots A^i$, $B^1 \dots B^j$, $D^1 \dots D^k$ and $T^1 \dots T^l$ and subsequent thermal treatment in the presence of a reactive or inert gas phase (tempering) or by common precipitation of sparingly soluble compounds and subsequent tempering or by loading of the support component $T^1 \dots T^l$ with salt solutions or gaseous compounds of the components $A^1 \dots A^i$, $B^1 \dots B^j$, $D^1 \dots D^k$ and subsequent tempering, wherein the employed salts are nitrates, sulfates, phosphates, carbonates, halogenides, oxalates, carboxylates, or mixtures thereof or carbonyl compounds or as acetyl acetonates.
10. The method according to claim 2, wherein the catalytic reaction is carried out with liquid, evaporated, or gaseous reactants.
11. The method according to claim 2, characterized in that the reactants for the catalytic reaction is supplied to several reactors and the product stream exiting the reactors is separately analyzed for each individual reactor.
12. The method according to claim 11, wherein for performing the catalytic reaction 5 to 1,000 reactors comprised of spaces with catalytically active material arranged therein are arranged parallel to one another or arranged in

arrays, wherein the diameter of these spaces is 100 μ m to 10 mm and the lengths are 1 mm to 100 mm.

5 13. The method according to claim 11, wherein, for a preset reactor length, the throughput of reactants is selected such that the desired degree of conversion is reached.

10 14. The method according to claim 11, wherein a monolithic block with several parallel channels, which can be closed selectively at the inlet or outlet side individually or in larger number also during the catalytic reaction, or a porous module having channels extending preferably parallel to the flow direction of the reaction mixture which channels can be selectively individually or in large number closed at the inlet or outlet side also during the catalytic reaction, is used as the reactor.

15. The method according to claim ¹⁶ ~~11~~, wherein the reactants for the catalytic reaction are ¹⁶ ~~supplied~~ supplied to the reactors ~~according to claim 11, 12, and 13,~~ and wherein the composition of the product streams exiting the reactors is analyzed by a measuring sensor, wherein the measuring sensor is guided two-dimensionally across the exit cross-section of all reactors or the reactors are moved two-dimensionally relative to the measuring sensor and the portion of the product streams received by the measuring sensor is supplied to the analytical device.

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